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<p>The final report describes the combined work of the Computational Chemistry and Aerothermodynamics branches within the Thermosciences Division at NASA Ames Research Center directed at understanding the signatures of shock-heated air. Considerable progress was made in determining accurate transition probabilities for the important band systems of NO that account for much of the emission in the ultraviolet region. Research carried out under this project has shown that in order to reproduce the observed radiation from the bow shock region of missiles in their boost phase it is necessary to include the Burnett terms in the constituent equation, account for the non-Boltzmann energy distribution, correctly model the NO formation and rotational excitation process, and use accurate transition probabilities for the NO band systems. This work has resulted in significant improvements in the computer code NEQAIR that models both the radiation and fluid dynamics in the shock region.</p>					
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Final Report for Boost-Phase Discrimination Research

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Foreword

This report describes work that was carried out in the Thermosciences Division at NASA Ames Research for the period 1990-1992. The work is the combined effort of the Computational Chemistry and Aerothermodynamics Branches. The principal goal of the work was the prediction of radiation signatures from shock layers from ascending vehicles. Some work was also directed at understanding the chemistry in the plume and plume induced separation. The research in the two branches was complementary in that both accurate molecular properties and the flow characteristics must be included in the codes that attempt to produce the signatures observed on actual flight experiments. Nevertheless, the work in the two branches is sufficiently distinct that the contributions from the two are described separately in the final report.

Computational Chemistry Branch

A. Statement of the Problem

The ultimate goal of the research is to be able to predict the radiation signature of ascending vehicles. This is essential in order to intercept missiles during their boost-phase. Our recent work is directed at demonstrating that detection of the hard body in the ultraviolet region where the Earth's background radiation is small is a viable concept. There are two actual flight experiments that have provided very accurate spectral data in the 3-5 km/s velocity range. In collaboration with the aerothermodynamics branch at Ames as well as other researchers in this program we have worked on understanding the physics of shock heated air and in the development of codes to predict the radiation signature as a function of altitude and speed. This section of the final report describes the work that was done in the Computational Chemistry Branch.

The molecular data that is needed to predict the bow-shock radiation and to understand the chemistry and emission from the plume can be divided into five major classes: (1) radiative transition probabilities for molecular transitions that contribute to the signatures; (2) transport properties such as thermal diffusion, thermal conductivity and viscosity needed to describe the flow in the shock; (3) thermodynamic properties such as heats of formation to understand the chemistry in the plume; (4) cross sections for electron molecule interactions; and (5) reaction rates for both equilibrium and non-equilibrium reactions.

One of the main objectives of our research program is to compute radiative intensity factors for air species that contribute to the radiation signatures of hard bodies and exhaust plumes. The NO molecule is one of the key species, as it is formed in significant concentrations in shock layers, for example, from the reaction of oxygen atoms with N_2 . During the three year period of this proposal we have determined accurate transition probabilities for the beta, ogawa, and epsilon systems of NO. The beta system ($B^2\Pi \rightarrow X^2\Pi$) was particularly challenging theoretically, because the upper state changes from Rydberg to valence character in the Franck-Condon region of the transition. Nevertheless, the radiative lifetimes as a function of vibrational level are in good agreement with recent laser induced fluorescence measurements. The theoretical results for the NO ogawa system ($b^4\Sigma^- \rightarrow a^4\Pi$) do not agree as well with experiment. However, we believe the theoretical values are the most accurate available for this band system.

More recently we have been computing accurate radiative intensity factors for the epsilon system of NO. The epsilon bands are a prominent constituent of shock-heated air in the spectral region between 180 and 210 nanometers. Our theoretical radiative lifetime of 19 nanoseconds for the low-lying vibrational levels is in good agreement with experiment. Thus our Einstein coefficients for this band system should represent a substantial improvement over existing values. These data are being incorporated into the NEQAIR code for computing the radiation from shock-heated air.

Emission from the Cameron band system of CO is observed in the plumes of rockets. To gain insight into the possible mechanism for this long-lived chemiluminescence, we have studied the quintet states of CO that are populated from recombination of ground state atoms. Of all the quintet states dissociating to ground state atoms, only two are found to be significantly bound. This work resulted in the assignment of two new band systems of CO, and it was shown that the emission observed between about 360 and 520 nanometers is due to the $a''^5\Pi \rightarrow a'^3\Sigma^+$ and $a''^5\Pi \rightarrow d^3\Delta$ transitions. Thus it is possible that the quintet states, populated through recombination of ground state atoms, serve as precursor states for the formation of the upper state of the Cameron system. However, the lifetime of this process is too short to explain the long-lived chemiluminescence from the Cameron system.

We computed the thermodynamic properties of various Al-Cl-O radical species that are expected to be present in rocket plumes. Specifically we determined the thermochemical heats of formation of plume species comprised of one or two aluminum atoms and various numbers of chlorine and oxygen atoms. Work is continuing on refining the geometry of the most stable isomer of Al_2O_2 with an extensive treatment of electron correlation.

Our laboratory has been a leader in the computation of accurate transport properties. During the last three years we considered the interaction of nitrogen and oxygen atoms and ions. The calculations are based on *ab initio* potentials and a semiclassical approximation to the collision integrals. This latter approximation has been shown to be valid at room temperature and above based on comparisons with an accurate quantum treatment of the scattering problem. These atom-atom and atom-ion transport properties such as thermal diffusion and viscosity are thought to be accurate to 5%. Calculations are in progress to determine accurate atom-molecule and molecule-molecule collision integrals for important air species.

Electron-molecule collisions become an important mechanism for exciting molecules at high velocities. These collisions are particularly efficient at scrambling the populations in the excited states. Thus to predict the emission from the shock layer, it is necessary to determine accurate cross sections for electron-molecule collisions between the excited states of important air species. In this proposal period we determined cross sections for some excited states of N_2 .

B Summary of Most Important Results

1. Accurate transition probabilities and branching ratios for the beta, gamma, and epsilon band systems of NO were determined. These are the most accurate presently available and these data have been incorporated into the NEQAIR codes used to predict the radiation from shocks.
2. Assigned several new band systems of the CO molecule involving transitions between the quintet and triplet states. Demonstrated that it is unlikely that the quintet states of CO could provide an excitation mechanism for the upper state of the Cameron band system observed prominently in plumes.
3. Accurate transport properties were computed for atom-atom and atom-ion interactions involving nitrogen and oxygen atoms. Collision integrals and transport properties were reported in a series of papers for a wide range of temperatures.
4. Electron collision cross sections were computed for some excited states of N₂, which are important in determining the state populations in the shock layer, especially at the higher velocities. We also computed vibrational excitation probabilities for N₂-N₂ collisions, which is a key mechanism for accommodating the energy in the shock layer.

C. List of Publications

Huo, W. M., *Electron Collision Cross Sections Involving Excited States, in Nonequilibrium Processes in Partially Ionized Gases*, M. Capitelli and J. N. Bardsley, editors, NATO Advanced Study Institute Series, p. 341 Plenum (1990).

Sharma, S. P., Huo, W. M., and Park, D., *Rate Parameters for Coupled Vibration-Dissociation in a Generalized SSH Approximation*, Journal of Thermophysics and Heat Transfer, **6**, 9 (1992).

Levin, E., Partridge, H., and Stallcop, J. R., *Collision integrals and high temperature transport properties for N-N, O-O and N-O*, J. Thermophysics and Heat Trans., Vol. 4, No.4, 1990, pp. 469-477.

Stallcop, J. R., Partridge H., and Levin, E., *Collision integrals for the interaction of the ions of nitrogen and oxygen in a plasma at high temperatures and pressures*, Physics of Fluids B -- Plasma Physics **4**, 386 (1992).

Stallcop, J. R., Partridge, H., and Levin, E., *Resonance charge transfer, transport cross sections, and collision integrals for $N^+(^3P)$ - $N(^4S^0)$ and $O^+(^4S^0)$ - $O(^3P)$ interactions*, J. Chem. Phys. **95**, 6429 (1991).

Partridge, H., Stallcop, J. R., and Levin, E., *Transport cross sections and collision integrals for $N(^4S^0)$ - $O^+(^4S^0)$ and $N^+(^3P)$ - $O(^3P)$ interactions*, Chem. Phys. Lett. **184**, 505 (1991).

Partridge, H., Langhoff, S. R., and Bauschlicher, C. W., *Theoretical study of the spectroscopy of NO^+* , J. Chem. Phys. **93**, 7179 (1990).

Langhoff, S. R., Partridge, H., Bauschlicher, C. W., and Komornicki, A., *Theoretical study of the NO beta system*, J. Chem. Phys. **94**, 6638 (1991).

Bauschlicher, C. W., Langhoff, S. R., and Partridge, H., *Theoretical study of the lowest $^5\Pi$ and $^5\Sigma^+$ states of CO*, J. Chem. Phys. **98**, 8785 (1993).

Bauschlicher, C. W., Partridge, H., and Langhoff S. R., *Theoretical study of the NO $b^4\Sigma^-$ and $O_2^+ b^4\Sigma_g^-$ radiative lifetimes*, Chem. Phys. Lett., in press.

D. List of All Participating Personnel

Stephen R. Langhoff
Charles W. Bauschlicher
Harry Partridge
Winifred Huo
James Stallcop
Eugene Levin
Andrew Komornicki

All personnel except James Stallcop are Ph. D. scientists- no advanced degrees were earned

Aerothermodynamics Branch

A. Statement of the Problem

The ultimate goal of the research within the Aerothermodynamics Branch is the same as that for Computational Chemistry Branch. The specific goal of Aerothermodynamics Branch is to develop a computer code that numerically reproduces the flight data. This consists of: (1) interpreting the flight data, (2) developing the theoretical model that describes the thermochemical phenomena occurring in the flow, (3) developing the CFD code that accommodates the thermochemical model, (4) run the code for the flight conditions, and compare the calculated results with the flight data.

Aerothermodynamics Branch has grants to both Stanford University, with Prof. Dean Chapman as the principal investigator, and North Carolina State University, with Prof. Graham Candler as the principal investigator, to carry out portions of the task. These two universities have had other professors, such as Prof. Robert MacCormack, and several graduate students to assist the principal investigators. Ames staff collaborated with these university personnel. In addition, Ames staff collaborated also with Institute for Defense Analysis (IDA), with Dr. Debrah Levin as the principal investigator, which has been funded independently of Ames.

The Bow Shock Ultraviolet Flight 1 was flown in April 1990, at a flight of about 3.5 km/s. The spectrometers and monochromators located in the nose region of the vehicle measured a substantial radiation in the ultraviolet region, which consists mostly of nitric oxide (NO) band systems. The Flight 2 was flown in February 1991 at a flight speed of about 5.1 km/s. In the second flight, the radiation from the plume region was determined as well as the nose region. One radiometer viewed the plume approximately along the centerline, while the other viewed far-field, that is, at 25 degrees off the centerline. The intensity of NO radiation in the nose region in the second flight behaved in the same as manner as in the first flight, though at a different level. Significant radiation was detected both from the centerline and the far-field regions of the plume.

B. Progress of the Work

Before the first flight, it has been predicted, based on existing experimental and theoretical data, that the flowfield will be in thermochemical nonequilibrium. After the first flight experiment, calculations were carried out for the radiative signal received in the blunt nose region of the vehicle using the then standard nonequilibrium codes. The standard codes consisted of the two-temperature nonequilibrium blunt body flow code and the nonequilibrium radiation code (NEQAIR). In the low altitude range, namely, between 40 and 50 km, the calculated radiation intensity agreed approximately with the flight data. However, at higher altitudes, the calculation underpredicted the measurement by at least seven orders of magnitude. This trend was later found to be the same even for the second flight.

This discrepancy between the measured and the calculated radiation intensity of NO set off concerted efforts within the Branch, IDA, and elsewhere. The immediate response was to attempt explaining the experimental data in terms of equilibrium, that is, that the flowfield and radiation mechanism were in thermal equilibrium. The equilibrium calculation yielded radiation intensity values that are uniformly an order of magnitude higher than the flight data at all altitudes. The fact that this discrepancy existed at altitudes of 40 to 50 km, where NEQAIR gave a satisfactory result, was particularly disturbing. This reinforced the belief that the flowfield is in nonequilibrium.

It was understood at once that the original version of the code NEQAIR is inappropriate for this problem because the intended flight speeds are different: the code assumes that electrons, prevalent only at high flight speeds (above 5 km/s), are responsible for excitation of internal states. Levin made a correction to the NEQAIR code to effect that it is the heavy particles, and not electrons, that cause excitation of electronic states. This correction raised the calculated radiation intensity at high altitudes by an order of magnitude, leaving the discrepancy to be two orders.

Thereupon, several hypotheses were developed to explain the still remaining discrepancy between the theory and the flight data. These included: (1) neglecting of Burnett terms in the constituency equations in fluid mechanical equations, (2) slip at the wall boundary, (3) inaccuracy in modelling of the rotational relaxation behavior, (4) inaccuracy in the value of the cross section for excitation of electronic states of NO, (5) inaccuracy in the spectroscopic and radiative parameters for NO band systems, (6) inaccuracy in modelling the NO formation processes, and (7) the nonBoltzmann distribution of translational mode in the shock wave.

Lumpkin et al first explored the effect of neglecting of Burnett terms. They found that inclusion of Burnett terms improved the theory slightly, that is, by an order of a factor of 1.5.

Lumpkin also pioneered modelling of the rotational relaxation correctly. This was done by extending the existing rotational model by incorporating the recent theoretical and experimental data obtained from the linewidth measurements. This correction produced a slight improvement. Later, Moreau et al made a drastic modification to rotational model by incorporating the so-called diffusion model. This correction greatly increased the calculated

radiation intensity.

The cross section for electronic excitation of NO molecule was studied experimentally at Stanford Research International (SRI). The work showed that the cross section is very large, that is, about 50 square angstroms.

Boyd et al explored the question of whether the non-Boltzmann energy distribution is responsible for the discrepancy between the flight data and the continuum calculations using the Direct Simulation Monte Carlo (DSMC) technique. It was shown first that DSMC technique is at least equivalent to the continuum approach in the regime where they are expected to be, and that indeed the non-Boltzmann distribution is partly responsible for the discrepancy. But the extent of this effect was found to be uncertain.

Laux et al and Moreau and Laux investigated the accuracy of the spectroscopic and radiation intensity parameters for NO by comparing the calculated results with the experiment conducted at Stanford University with a plasma torch. By using the spectroscopic and radiative parameters generated recently at Computational Chemistry Branch, rather than the set used in the original version of NEQAIR, they were able to demonstrate that the experimental data can be closer reproduced. This led to creation of the NEQAIR-2 code. Use of this code narrowed the discrepancy still further.

Moreau developed a five-temperature model to describe the process of formation and removal of NO molecules in an experiment conducted at CALSPAN (W. Wurster's experiment). The model so developed can numerically recreate CALSPAN experiment by assigning a vibrational temperature of NO molecule which is different from that of N₂ molecule. The new model lowers the calculated radiation intensity at high altitudes.

The last two innovations by Moreau have not yet been fully incorporated in the calculation of radiation intensity. However, if one assumes that the cross-section of excitation of NO electronic states is infinitely large, then, by incorporating all the improvements made, the calculated intensity agrees closely with the flight data at high altitudes, for the first flight. For the second flight, the same approaches overestimates the radiation intensity.

In order to study the dynamics of the plume, a theoretical calculation was first made of the fluid flow in the plume. The flight data on the plume region were calculated and compared by Candler et al. A problem exists in that the thermochemical state of the rocket effluent at the nozzle exit is uncertain. By adjusting the state parameters at the exit, it was shown that the observed radiation intensity along the centerline can be numerically recreated. The radiation is attributed to that from the solid particulates in the effluent. However, for the far-field radiometer, that is, that viewing at 25 degrees, the calculation severely underpredicted the measured value. The experimental data can be recreated numerically by first assuming that the radiation emanates mostly from the CO Cameron band system, and that the relative populations of electronic states of CO molecules are frozen at the nozzle exit conditions. Thus, the far-field radiation signal seems to emanate from the gas-phase radiation.

C. Summary of Most Important Results

1. It was shown that, by including the Burnett terms in the constituent equation, accounting for the non-Boltzmann energy distribution, correctly modelling the NO formation and rotational excitation processes, and using the correct transition probabilities for NO radiation, one can numerically reproduce the observed radiation of NO in the bow shock region for the flight velocity of 3.5 km/s, provided the rate of excitation of the electronic states through the vibration-electronic energy transfer is very fast. (For the 5 km/s flight, the agreement between calculation and measurement is not as good.)
2. It was found that the DSMC technique is at least equivalent to the continuum technique in determination of nonequilibrium radiation.
3. In the expanding plume flow behind the second flight vehicle, the electronic excitation temperature seems to be frozen at the value at the exit of the nozzle.

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Boyd, I. D., and Gokcen, T., "Assessment of Thermochemical Models for Continuum and Particle Simulations of Hypersonic Flow," AIAA Paper 92-2454, July 1992.

Boyd, I. D., and Gokcen, T., "Computation of Axisymmetric and Ionized Flows Using Particle and Continuum Methods," AIAA Paper No. 93-0729.

Candler, G. V., Levin, D. A., Collins, R. J., Erdman, P. W., Ziph, E., and Howlett, C., "Comparison of Theory with Plume Radiance Measurements from the Bow Shock Ultraviolet 2 Rocket Flight," AIAA Paper 92-0125, to be published in Journal of Thermophysics and Heat Transfer.

Candler, G. V., Boyd, I., Levin, D. A., Moreau, S., and Erdman, P., "Continuum and DSMC Analysis of Bow Shock Flight Experiments," AIAA Paper 93-0275, January 1993.

Candler, G. V., Levin, D. A., Walberg, G., and Howlett, C., "Assessment of Small Rocket Flight Experiments to Obtain Nonequilibrium Radiation Data," AIAA Paper 93-0143, January 1993.

Erdman, P. W., Ziph, E. C., Espy, P., Howlett, C., Levin, D. A., Loda, R., and Candler, G. V., "Flight Measurements of Low Velocity Bow Shock ultraviolet Radiation," AIAA Paper 91-1410, June 1991.

Erdman, P. W., Ziph, E., Espy, P., Howlett, C., Levin, D. A., and Candler, G. V., "In-Situ Measurements of UV and VUV Radiation from a Rocket Plume and Re-entry Bow Shock," AIAA Paper 92-0124, January 1992, (to be published in Journal of Thermophysics and Heat Transfer.)

Erdman, P. W., Espy, P., Levin, D. A., and Candler, G. V., "Measurements of Ultraviolet Radiation From a 5 km/sec Bow Shock," AIAA Paper 92-2870.

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Lumpkin, F., "Development and Evaluation of Continuum Models for Translational-Rotational Nonequilibrium," Ph. D. Thesis, Stanford University, 1990.

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Moreau, S., Chapman, D. R., and MacCormack, R. W., "Effect of Rotational Relaxation and Approximate Burnett Terms on Hypersonic Flow-Field Radiation at High Altitudes," AIAA Paper 91-1702.

Moreau, S., Bourquin, P. Y., Chapman, D. R., and MacCormack, R., "Numerical Simulation of Sharma's Shock Tube Experiment," AIAA Paper 93-0273.

Moreau, S., and Laux, C., "A More Accurate Nonequilibrium Air Radiation Code: NEQAIR Second Generation," AIAA Paper No. 92-2968, July 1992.

Moreau, S. "Computation of High Altitude Hypersonic Flow Field Radiation," Ph.D. thesis, Stanford University, 1993.

Venkatapathy, E., and Feiereisen, W. J., "Computational Analysis of Plume Induced Separation," AIAA Paper 91-0711, January 1991.

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North Carolina State University: Graham Candler and Brian Landrum.